Organic Electrochemical Transistors

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Abstract

Organic electrochemical transistors (OECTs) leverage ion injection from an electrolyte into an organic semiconductor film to yield compelling advances in biological interfacing, printed logic circuitry and neuromorphic devices. Their defining characteristic is the coupling between electronic and ionic charges within the volume of an organic film. In this review we discuss the mechanism of operation and the

materials that are being used, overview the various form factors, fabrication technologies and proposed applications, and take a critical look at the future of OECT research and development.

Introduction

The invention of the transistor in 1947 heralded the era of microelectronics. Made of semiconductors and metals, it enabled the control of the flow of electrons in a solid-state device, providing thereby a more robust alternative to vacuum tubes. Its miniaturization led to the development of the integrated circuit, which is at the heart of every modern electronic device. Modern transistors rely on field-effect doping for their operation: The number of mobile electrons (*n*-type) or holes (*p*-type) inside a semiconductor is modulated by a voltage applied to a metallic electrode that is separated from the semiconductor by a thin insulating layer.[1] The appropriately descriptive term MOSFET, short for metal/oxide/semiconductor field-effect transistor, is used to refer to these devices. Interest in organic electronic materials, and in particular in their potential for low cost fabrication over large areas, led to the development of organic field-effect transistors (OFETs).[2] These devices utilize conjugated (semiconducting) small molecules and polymers, and offer an alternative to inorganic devices in applications where facile processing on various substrates and tunability of electronic properties are required. Steady progress in the past three decades has led to exciting demonstrators ranging from pixel drivers for displays[3] to bionic skin.[4]

The organic electrochemical transistor (OECT) was developed by Wrighton and colleagues in the mid-eighties.[5] The typical structure of an OECT is shown in **Fig. 1a**. It consists of an organic semiconductor film which is in contact with an electrolyte, in which an electrode (the gate) is immersed. Metal electrodes, the source and drain, establish contact to the organic semiconductor film and define the channel, in which holes or electrons flow from the source to the drain. The operation of an OECT relies on ions which are injected from the electrolyte into the organic film, change the doping state, and hence the conductivity, of the latter.[6] The operation, which has been described using both the language of electrochemistry and solid state physics, is controlled by voltages applied to the gate (gate voltage, V_g)

and to the drain (drain voltage, V_d), which are referenced with respect to the source electrode. The gate voltage controls the injection of ions into the channel, hence controls the doping state (redox state in the language of electrochemistry) of the organic film. The drain voltage induces a current (drain current, I_d) which is proportional to the quantity of mobile holes or electrons in the channel and therefore probes the doping state of the organic film. Like their MOSFET and OFET predecessors, the OECT can be viewed as a switch, in which the gate voltage (input) controls the drain current (output). It can also be viewed as an amplifier, in which the power of a signal applied at the input comes out amplified at the output.[7]

A prototypical material for OECTs is the conducting polymer poly(3,4-ethylenedioxythiophene) doped with poly(styrene sulfonate) (PEDOT:PSS). In this material, the semiconducting PEDOT is doped *p*-type (oxidized in the language of electrochemistry), leading to mobile holes that can hop from one chain to the other, giving rise to a hole current when a drain voltage is applied. These holes are compensated by sulfonate anions of the PSS (which play the role of ionized acceptors in the language of solid-state physics).[8] OECTs based on conducting polymers such as PEDOT:PSS work in depletion mode: In the absence of a gate voltage, a hole current flows in the channel (ON state). When a positive gate bias is applied, cations from the electrolyte are injected in the channel and compensate the anions (**Fig. 1b**). This is equivalent to compensation doping, i.e. implanting donors in *p*-type silicon. As a result, the number of holes in the channel decreases (holes extracted at the drain are not replenished at the source) and the film is dedoped. This leads to a drop in the drain current, and the device reaches the OFF state.[6] Accumulation mode OECTs have also been reported. These devices are made of semiconducting polymers (**Fig. 1c**), and the transistor is normally in the OFF state due to the small number of mobile holes in the channel. Application of a negative gate voltage causes injection of anions into the channel and a corresponding accumulation of holes (electrochemical doping in the language of chemistry), leading to the ON state.[9]

The identifying characteristic of OECTs is that the doping changes occur in the whole volume of the channel, as opposed to a thin interfacial region of field-effect transistors (see **BOX 1**). As we will see below, this means that large modulations in the drain current can be achieved for low gate voltages and this makes OECTs efficient switches and powerful amplifiers.[7,10] This has triggered interest in a variety of applications including neural interfaces,[11,12] chemical and biological sensors,[13,14] printed circuits,[15,16] and neuromorphic devices.[17,18] The use of electrolytes instead of MOS capacitors allows large flexibility in device architecture, and unlocks the potential for integration with a variety of substrates, taking on unique form factors, and using a broad range of fabrication processes. The inherent tunability of organic molecules permits optimization of ionic and electronic transport and enables facile biofunctionalization. All these factors make OECTs a device offering unique potential for a wealth of applications. In the paragraphs that follow we discuss the physics of OECTs, review the different materials that are being used, discuss the evolution of their technology, highlight the different applications proposed, and take a critical look at the future of OECT research and development.

Physics

OECTs transduce small voltage signals applied to the gate into large changes in the drain current, and this makes them useful for a variety of applications. The transduction process is described by the transfer curve, which shows the dependence of the drain current on the gate voltage (**Fig. 1b,c**). The steeper the transfer curve, the larger the change in drain current for a given gate voltage signal. The efficiency of the transduction process is measured by the first derivative of the transfer curve which is the transconductance, $g_m=\partial I_d/\partial V_g$, an important figure-of-merit for transistors. OECTs have been shown to possess very large transconductance values (of the order of mS for micron scale devices),[7] a fact that is attributed to the volumetric nature of their response. Bernards et. al., developed a simple model that

captures this essential piece of physics of OECTs, namely, the fact that ions from the electrolyte enter the channel and change the electronic conductivity throughout its volume.[6] According to this model, the device is divided into two circuits, the ionic one, which treats the flow of ions in the gate/electrolyte/channel structure, and the electronic one, which describes the flow of electronic charge in the source/channel/drain structure according to Ohm's law (**Fig. 1d**). The electronic circuit is therefore treated as a resistor, in which electronic charge drifts under the influence of the local potential, in an identical fashion as in MOSFETs. The ionic circuit, on the other hand, consists of a resistor, describing the flow of ions in the electrolyte, in series with a capacitor, describing the storage of ions in the channel (the capacitance of the gate/electrolyte interface is assumed to be negligible). This implies a purely capacitive process, according to which ions injected in the channel do not exchange charge with the organic film, but rather electrostatically compensate the presence of charges of opposite sign.[19] There are no electrochemical reactions between the electrolyte and the channel in this model.

The Bernards model provides a good fit for the output characteristics of OECTs and makes quantitative predictions for the transconductance. In saturation, and for depletion mode devices, it gives:[12]

$$g_m = (W/L)\cdot d\cdot\mu\cdot C^*\cdot(V_{th}-V_g), \qquad (1)$$

where W, L and d stand for channel width, length, and thickness, μ is the mobility of the electronic carriers, C^* is the capacitance per unit volume of the channel, and V_{th} is a threshold voltage (for accumulation mode devices the voltage terms are reversed). This equation is similar to the one for field-effect transistors, the only difference being that the product $d\cdot C^*$ replaces the capacitance per unit area of the MOS capacitor, C', of FETs. This observation is at the heart of the difference between the two devices. While in an FET the physical thickness of the channel does not enter in this equation, in OECTs channel thickness is a parameter that can be used to tune performance. An example illustrates this point

nicely: In a typical OFET with a 100 nm thick SiO_2 oxide, C' is in the tens of nF/cm^2.[20] In OFETs gated with an electrolyte (see **BOX 1**), C' is that of the electrical double layer, of the order of 1-10 μF/cm^2.[21,22] In an OECT with a 130 nm thick PEDOT:PSS channel, the product of $d \cdot C^*$, which is the equivalent capacitance per unit area, is 500 μF/cm^2.[12] Volumetric gating, therefore, gives OECTs a performance edge in terms of amplification compared to other transistor technologies.[7]

The high transconductance of OECTs comes at a cost of rather slow operation. The Bernards model predicts that the response time can either be limited by the ionic circuit or by the electronic one.[6] In most practical devices it is the ionic circuit that dominates and the response time, which is dictated by the product of the resistance of the electrolyte and the capacitance of the channel. The latter is proportional to channel thickness meaning that OECTs become slower as d increases. The ramification is that channel thickness can be used as parameter to adjust the tradeoff between gain and bandwidth.[12] Practically speaking, microfabricated OECTs with liquid electrolytes showing response times as fast as a few tens of μs have been reported.[7] Such speeds limit the scope of application of OECTs to applications requiring maximum frequencies in the tens of kHz range. This is adequate for most biosensor applications, which are quasi-static, as well as for recording electrophysiological signals.[23] The use of gel or solid electrolytes leads to slower OECTs, yet there is plenty of room for applications in areas such as printed electronics for these devices.[24]

The Bernards model offers a good starting point for understanding the basic physics of OECTs, but there are other considerations that have been or still need to be taken into account. Spatially-resolved voltage measurements along an OECT channel, for example, imply that the conductivity along the channel varies in a non-linear fashion with charge density.[25] Introducing a charge density-dependent conductivity in the Bernards model was shown to improve the quality of fits to the output curves of PEDOT:PSS OECTs.[26] Issues with non-ideal contacts were also identified in OECTs.[27] Finally, OECTs represent the limit where

ions penetrate freely in the volume of the channel (electrolyte-gated FETs represent the other limit, where ions accumulate at the surface of the channel). Several examples of such devices have been reported in literature, using materials that show a capacitance that is proportional to film thickness.[9,12,28] In certain cases, behavior between the two limits was observed,[29,30] hinting to the presence of a barrier for ion injection. These results point to an important fundamental issue: While the physics of hole and electron injection and transport in organic semiconductors has been studied extensively in the past few decades, little is known about the processes of ion injection and transport in organic semiconductors. We discuss this topic further in the final section.

The fact that OECTs are electrolyte-gated devices introduces some particularities in their device physics. One of these is that the fraction of the applied gate voltage that drops on the channel is controlled by the nature and the geometry of the gate electrode. If a polarizable electrode such as Pt or Au is used as the gate, two capacitors are formed in the ionic circuit: one corresponding to the electrical double layer formed at the gate/electrolyte interface, and another corresponding to the volumetric capacitance of the channel. Since these capacitors are in series, the applied gate voltage drops on the smaller one (**Fig. 1e**). For efficient gating, the area of the gate electrode must be much larger than that of the channel, otherwise a large fraction of the applied gate voltage will drop at the gate/electrolyte interface. Such large gates might be difficult to accommodate in some applications, however, using a thick PEDOT:PSS electrode as the gate can help achieve a large gate capacitance. An alternative option is to use a non-polarizable gate electrode such as Ag/AgCl. In this case the voltage drop at the gate/electrolyte interface is negligible,[31] and effective gating is achieved. It should be noted, however, that in some sensing applications, a small gate electrode is preferable, for example when a sensing reaction is meant to occur at the gate, and the channel merely acts as the transducer.[32] Finally, the nature (liquid, gel, solid) and ion concentration of the electrolyte influence the response time of the transistor, as electrolyte conductivity determines the resistor of the ionic circuit.[33]

Materials

The channel in the vast majority of OECTs reported in literature is made of a conducting polymer (based on PEDOT or polypyrrole, doped *p*-type with small anions or polyanions, **Fig. 2a,b**). These materials are synthesized by solution-, vapor phase-, or electrochemical polymerization. By far the most popular material is PEDOT:PSS, which, over the last 15 years, has cemented its status as the "fruit fly" in OECT literature. This is largely due to its commercial availability in the form of aqueous dispersions,[34,35] which permit the facile deposition of thin films using solution processing techniques. These dispersions are prepared by polymerizing EDOT (the monomer of PEDOT) in the presence of PSS. Films of PEDOT:PSS cast from these dispersions show high electronic[8] (hole) conductivity and are widely used in electrostatic coatings and as anodes for light emitting diodes and solar cells.[8] As a result, their structure and electronic conductivity have been the subject of numerous investigations.[36,37] Various co-solvents, surfactants, and processing methods have been used to maximize their hole conductivity, currently in excess of 1,000 S/cm.[38] Their ionic conductivity, on the other hand, is a topic of more recent investigations.[39–41] Finally, PEDOT:PSS films are compatible with biological milieu as they show good electrochemical stability in aqueous electrolytes,[42] and can be rendered insoluble in water through the addition of crosslinkers such as (3-glycidyloxypropyl)trimethoxysilane (GOPS),[43,44] and divinylsulfone (DVS).[45] Overall, it is rather simple to make PEDOT:PSS OECTs with a transconductance in the range of mS and a response time in the range of tens of μs,[7] which makes these devices attractive for a variety of applications discussed below.

Despite its many attractive properties and high performance in OECTs, PEDOT:PSS suffers from some obvious limitations. Its structure is complex along a broad range of length scales, which limits its use as a model system for the development of structure vs. properties relationships. In terms of electrical

properties, the bulky structure of PSS limits the volume fraction of PEDOT in the film, and hence limits the volumetric capacitance.[19] Moreover, conjugated polymer motifs with higher hole mobility than PEDOT have been developed.[46] In terms of mechanical properties, the Young's modulus of PEDOT:PSS is considerably higher than that of most biological tissues,[43,47] which can be a limitation for applications in bioelectronics. In terms of biological properties, while PEDOT:PSS films are broadly cytocompatible,[48] polymers or composites allowing more versatile biofunctionalization would be desirable. Finally, it terms of processing, the acidity of PSS can cause corrosion of print heads. These limitations are motivating the synthesis of new materials for OECTs (including those in **Fig. 2c-f**).

One approach for new OECT materials involves replacing the PSS with less acidic polyanions, such as polymers comprising (trifluoromethylsulfonyl)imide side groups. OECTs made from these materials were shown to have similar performance in terms of transconductance and response time to that of PEDOT:PSS-based OECTs[49] A second approach involves replacing PSS altogether by attaching ionic groups through side chains directly on the thiophene backbone. Such conjugated polyelectrolytes can be semiconducting when the ion is compensated by a counter ion (**Fig. 2c**), or conducting, when it is compensated by an electronic charge on the conjugated backbone (**Fig. 2d**). One example is a polythiophene with a sulfonate group attached on the backbone with a hexyl chain; a semiconductor that gave rise to accumulation mode OECTs with transconductance of the order of mS.[50] Another example is a PEDOT with a pendant sulfonate group, a conductor that gives rise to depletion mode OECTs.[51] Blending a semiconducting and a conducting conjugated polyelectrolyte yields OECTs whose characteristics can be tuned by balancing the stoichiometry of the channel.[51] A third approach to OECT materials involves the use of hydrophilic or ion transporting side chains on conjugated polymers. OECTs from p(g2T-TT), a polythiophene with glycolated side chains (**Fig. 2e**) exhibited transconductance values outpacing these of PEDOT:PSS-based OECTs of the same geometry.[30] Using the same type of side chain functionalization, an *n*-type copolymer p(gNDI-g2T), based on naphthalene-1,4,5,8-tetracarboxylic

diimide and bithiophene units (**Fig. 2f**), was developed and ambipolar OECTs made thereof exhibited high stability during pulsed measurements over 2 h in aqueous media.[28] Finally, a fourth approach we wish to highlight here is the blending of PEDOT:PSS with poly(vinyl alcohol) (PVA). The latter provides a handle for subsequent silanization, allowing for covalent linkage of biological moieties onto the films without any deleterious effects on electrical properties.[52] Functionalization with polypeptides and proteins, shown to maintain their biological functionality, was demonstrated.

Although electrochemical polymerization was used to fabricate the first OECT,[5] and despite the fact that it provides a versatile way of incorporating biomolecules in conducting polymers,[53] it has fallen out of favor. This is probably due to the fact that this polymerization method relies on growth from a conducting surface, which is hard to implement in OECTs. Vapor phase polymerization (VPP), on the other hand, seems to be a promising way for incorporating different biomolecules in the channel of OECTs. According to this process, a film cast from a precursor solution containing tosylate (TOS, the pendant group of PSS) moieties is exposed to vapor of EDOT to yield the conducting polymer PEDOT:TOS (Fig. 2b).[54] Mixing different polymers in the precursor solution provides a facile way to make high quality composites. Indeed, composites with poly(ethylene glycol) (PEG) showed similar OECT performance as pristine PEDOT:TOS.[55] A carboxyl-containing PEG was also used, showcasing the potential of VPP in applications requiring biofunctionalization. Furthermore, gelatin, a derivative of the extracellular matrix protein collagen, was incorporated into PEDOT:TOS. This did not change the electrochemical properties of the conducting polymer, nor its performance in OECTs.[56] These composites were found to specifically support bovine brain capillary endothelial cell adhesion and growth, indicating that the functionality of the biomolecule was maintained. Finally, VPP was used to yield composites with poly(tetrahydrofuran) that exhibited a hysteresis effect in their OECT characteristics.[57] The hysteresis was attributed to a conformational change of the polymer structure during redox cycling and was used to demonstrate a non-volatile memory device.

A variety of electrolytes including gels,[58] and solids,[59] and a variety of gate electrodes including traditional polarizable (such as Pt)[60] and non-polarizable electrodes (such as Ag/AgCl),[60] as well as PEDOT:PSS[59] and various forms of carbon[61,62] have been used to fabricate OECTs. The specific choices for these materials are often dictated by the application needs or limitations imposed by fabrication. For testing new channel materials, the use of pre-patterned Au source and drain electrodes on glass substrates, Ag/AgCl gate electrodes, and aqueous solutions of NaCl or KCl is widespread. Key fabrication issues and application specific requirements are reviewed in the following sections.

Technology

Already in 1980, Diaz and Castillo reported indirect measurements of the reversible electrochemical switching of electronic conductivity in polypyrrole (PPy), recorded from films made on Pt electrodes.[63] When Wrighton and co-workers electropolymerized PPy onto Au separated by 1.4 µm gaps on SiO_2-coated Si-wafers, they found that PPy thick films could extend and even bridge the gaps in between the electrodes.[5] Using a two-circuit drain-source and gate-counter/reference electrode probing approach, the authors could record clear accumulation-mode transistor action in the resulting devices. With similar structures and different probing set-ups, the authors used PPy, poly(3-methylthiophene)[64] and polyaniline[65] as the active layer in electrochemical transistors for redox sensing and "chemiresistors" applications.[66] From 1985 to 2000, several demonstrations of organic electrochemical transistors were reported, still based on microfabricated lateral electrode structures made on rigid substrates, targeting (i) development of a sandwich structures,[67] (ii) incorporation of solid electrolytes,[68,69] (iii) fabrication of pyrrole-N-methylpyrrole transistor channels that incorporate the diaphorase enzyme,[70] (iv) advancement towards high performance in aqueous and non-aqueous electrolytes.[71]

The use of electrolytes as gating media allows a great deal of flexibility in their fabrication of OECTs in terms of the placement of the gate electrode with respect to the channel. Moreover, OECTs do not suffer from "long-channel" effects, since the entire channel volume contributes to the conduction of the transistor-modulated current. Together these facts enable OECTs with low operating voltage to be designed using very relaxed criteria with respect to gate, electrolyte and channel dimensions and relative positions. The possibility, therefore, enables the use of various deposition and patterning techniques that allow the manufacturing of OECTs on a variety of substrates, and paves the way for radically new device architectures and form factors (**Fig. 3**). In 1994, the first electrochemical transistor was reported that in part was manufactured using printing techniques. Carbon-based drain and source electrodes, and dielectric insulators were screen-printed on a PVC base and the stack was then cracked in liquid nitrogen. Along the fractured edge, polyaniline was anodically grown, then coated with glucose oxidase (GOx) immobilized in poly(1,2-diamonibenzene). The resulting device, with a channel length and width of 20 µm and 4.5 mm, respectively, was explored as a microelectrochemical enzyme transistor for glucose sensing,[72] an approach later on also explored for peroxide sensing.[73] An OECT with a channel length of 0.5 mm gated through a thick filter paper soaked with 1M KCl electrolyte was also reported in 1998.[74]

In the late 1980s, PEDOT[75,76] was developed and later on also explored as electronic inks and coatings for a wide range of conductive, electronic and electrochemical applications.[77] In conjunction with PSS as the dopant, it showed good redox stability and high electronic conductivity. These combined features, promoted the development of high-performing and novel OECT architectures, where PEDOT:PSS serves both as the active channel and as the electrode for gate, drain and source, manufactured using for instance printing technology. In the late 1990s and early 2000s, a few attempts were conducted to study and explore the *in-situ* redox-switching of the electronic conductivity in PEDOT using different electrochemical setups and device configurations.[78,79] In 2002, the first demonstrations of using PEDOT

in transistor structures were reported.[59,80,81] In one example, by deactivating/patterning PEDOT:PSS coatings, using screen-printing, made it possible to form an all-laterally configured gate, drain and source OECT with a screen-printed gelled electrolyte on top.[80] In such lateral OECTs, drain current ON/OFF ratios of 10^5, a transconductance of 1.2 mS, and a maximum frequency operation of 200 Hz were achieved.

Sparked by the liberation in terms of easy manufacturing protocols to achieve high-performing and low-voltage operating OECTs, further developments of various novel OECT-structures, based on PEDOT:PSS and other mixed ionic/electronic polymer conductors, and associated high-volume manufacturing processes were investigated. Lateral PEDOT:PSS-based OECTs were combined with vertical electrochromic display cells, all printed-manufactured on coated paper, to form actively addressed smart pixels and displays.[82] Long PEDOT:PSS channels enabled the integration and gating via microfluidic systems made from polydimethylsiloxane (PDMS),[83] a technology pathway that later on resulted in OECT-gating through a phospholipid membrane,[84] and later through cells.[85] PANI, PPy and PEDOT nano-wires were grown in between Pt source and drain electrodes and remotely gated via an electrolyte.[86]

Paper and plastic foils, being the largest surfaces ever manufactured, have attracted great attention from the OECT community. Several attempts to utilize standard and modified printing techniques have been explored to produce OECT devices and circuits along those surfaces, such as using screen-printing,[87] and inkjet printing.[88] All-screen-printed OECT logic circuits, such as flip-flops and shift registers operating at 1.5 V, was achieved on poly(ethylene terephtalate) (PET) substrates.[16] An ultra-thin carrier, made on a 4 µm thin parylene sheet, with OECTs has recently been explored in bioelectronics applications.[11] Textiles are yet another attractive carrier for OECTs, since it opens up for applications in wearable electronics. For instance, PEDOT-based transistors has been made on Gore-Tex, to enable gas sensors on “breathable” substrates,[89] and was also screen-printed on common fabrics to

form a washable sensors for biomarker determination, targeting sensing of external biological fluids (e. g. sweat, saliva and urine).[90]

Paper and textiles are built up from fibers and several attempts have been carried out to achieve OECT devices and even complex circuits on individual or combinations of fibers.[91] OECTs based on PPy made on a nylon nanofiber were recently explored for lead ion detection,[92] and PEDOT:PSS was soaked into natural cotton fibers to form the OECT channel for saline sensing.[93] PEDOT:PSS has also been explored in combination with nano-fibrillated cellulose (NFC) for OECT applications. As the cladding layer around NFC fibers, a scalable technology for self-standing and large-scale OECT systems is at hand. For instance, a reconfigurable OECT stickers platform was demonstrated,[94] and a transconductance beyond 1 S was recently demonstrated in PEDOT:PSS-NFC OECTs.[95] Finally, a unique form factor was achieved by making OECTs from PEDOT derivatives that were infused inside living plants.[96] Such OECTs could, in the future, be used to monitor and control plant development.

Applications

Bioelectronics

A major contemporary research area involves the use of organic electronic materials in bioelectronics,[48,97,98] with the aim of providing novel/improved tools for healthcare and biomedical research. OECTs are already playing an important role in this arena. On example is in electrophysiology, where they are used to interface with electrically active tissues and organs (**Fig. 4a**). Microfabricated PEDOT:PSS OECTs on parylene substrates were placed on the surface of the brain of rat models of epilepsy and shown to record seizures with a signal-to-noise ratio that was over 20 dB higher than that of PEDOT:PSS electrodes.[11] They were also shown to record activity arising from deeper in the brain,

improvements that were attributed to local signal amplification. Leveraging the direct, oxide-free contact of the conducting channel with cerebrospinal fluid, OECTs were used to inject current and stimulate neurons in a localized fashion.[99] OECTs were further integrated with organic thin film transistors to make arrays capable of recording myograms in transgenic rats with high temporal resolution.[100] Cutaneous applications are also being explored, with OECTs demonstrated to record cardiac activity from the arm of a human volunteer.[101] They were also used as amplifiers of recordings of human electrophysiological signals from the brain, heart, and muscle.[12,102,103] Finally, PEDOT:PSS OECTs were shown to be sterilizable by autoclaving, paving the way for clinical applications.[104]

Another application of OECTs in bioelectronics involves their use in conjunction with cell cultures (**Fig. 4b**). A major driver for this is the need for development of in vitro models that substitute animal use in toxicology and drug screening. OECTs have been used to record action potentials from cardiomyocytes with excellent signal-to-noise ratio,[105] and OECT arrays provided a spatial map of the electrophysiological activity.[106] A great deal of attention has been paid to non-electrogenic cells, where OECTs have been shown to monitor cell coverage[107] as well as barrier tissue formation and health.[108–112] The majority of these measurements involve the growth of a cell monolayer between the channel and the gate, which introduces an extra barrier for ion motion in the electrolyte and alters the characteristics of the OECT.[113] The same principle was applied to study ion channels in supported lipid bilayers assembled on PEDOT:PSS channels.[114] Sophisticated measurement protocols using ac modulations[115] or white noise[116] applied at the gate were developed and OECTs were shown to have superior performance to impedance sensing using electrodes. Given the optical transparency of PEDOT:PSS in the visible part of the spectrum, a key advantage of these devices is that they permit simultaneous optical and electronic readouts.[117] A powerful demonstration of the use of OECTs integrated with epithelial cell layers was the detection of enteric pathogens in milk.[118] Following the trend to transition from 2D to 3D cell culture models, OECTs were interfaced with cysts and shown to

measure changes occurring after assault by toxic compounds.[119] Finally, it should be mentioned that the arrangement of epithelial cells on an OECT channel can be controlled by the applied drain and gate voltage.[120] This offers unique opportunities to not only monitor, but also control cell behavior with these devices. To this end, OECTs have even been fabricated using 3D porous sponges of PEDOT:PSS, with potential future applications for hosting and monitoring cells.[121]

OECTs are also being used as transducers in biosensors (**Fig. 4c**). One application is human health and performance monitoring, which involves the detection of electrolytes (levels of potassium, sodium, etc). and metabolites such as glucose and lactate. For the detection of ions, a variety of approaches to gain selectivity have been developed.[122,123] For the detection of metabolites, the principle of sensor operation relies on the use of the appropriate redox enzyme that interacts selectively with the metabolite of interest and subsequently transfers an electron to the gate of the OECT. This was shown to change the drain current in a fashion that depends on the concentration of the metabolite.[32] Following early work where the enzyme was entrapped in the channel[72] or dissolved in the electrolyte,[124] immobilization on the gate electrode led to highly selective and sensitive sensors.[125,126] The integration of OECTs with microfluidics paves the way for multi-analyte sensor platforms,[127,128] while their integration with textiles envisions their use in wearable applications. i.e. detection of analytes in sweat.[90,129] Although the bulk of the work focused on detection in saline solutions, examples of detection in breath,[130] sweat,[131] and cell culture media[132] were also shown. The latter is a demonstration of the potential of OECT-based sensors for translation to the clinic, for distinguishing metastatic potential in tumor cell cultures.[132] Finally, OECTs were shown to provide an easy way to amplify fast scan cyclic voltammetry, allowing for much simpler instrumentation.[133] They were shown to enable selective detection of dopamine in the presence of interfering compounds, with sensitivities and limits of detection comparable or even better than those obtained using sophisticated electrochemistry techniques.[134]

Circuits and logic

The high transconductance and ON/OFF values along with low-voltage operation have encouraged scientists to explore OECTs also in circuits (**Fig. 4d**). Though, their relatively low switching speed compared with MOSFET technology typically excludes them from applications in (digital) signal processing and computation. However, combining OECTs with silicon-based MOSFETs creates many promising opportunities, for example in providing internet connectivity in electronic label and skin patch applications.[135] A first step in demonstrating operational OECT circuits was taken in 2002 when OECTs were included as pixel drivers in an active-matrix addressed electrochromic display manufactured on paper.[82] Further development to improve the matrix display fill factor[136] and to achieve digital NAND and NOR gates,[137] based on similar PEDOT:PSS OECT configurations, were later on also reported. Textiles and wire configurations represent another unique large-area and flexible platform for OECTs, besides paper and plastic foils, where potentially also the fiber grid can serve as a template, or even as an integral part of devices, to connect and define circuits. Woven OECT logics[91] was presented already in 2007 and functional NOR gates and inverters, using parallel PEDOT:PSS-based fabric wires combined with a polyelectrolyte, have also been achieved.[138] As with display pixels, sensor and detector devices can also be arranged in matrices to enable spatially resolved sensing or detection. OECTs can here serve as the addressing switch or line and row driver to enable x-y addressing of individual sensors in a cross-point matrix. In an effort to achieve a touch-less control interface, OECTs were printed and integrated to form addressing columns and read-out rows for a ferroelectric sensor matrix.[139]

Various analogue and logic OECT circuits have been realized using different large-area and high-volume manufacturing techniques, such as screen-printing,[16,140] lamination,[136] nanoimprint,[141] and combined inkjet printing and vapor-phase polymerization.[142] As the OECT circuit technology gradually has matured during the last decade, dedicated circuits for vastly different applications have been reported. With a

PEDOT:PSS-based signal-ON sensor circuit a ten-fold increase in sensitivity, as compared with performing sensing with only one OECT sensor, was reported.[143] Recently, the concept was extended to a Wheatstone bridge consisting of two PEDOT:PSS-based OECTs and used for lactate sensing.[132] A voltage amplifier, combining a high-transconductance OECT with a resistor, resulted in improved recordings of electrocardiographic signals.[103] Another potential area for electrolyte-based devices is the supercapacitor technology and energy storage in general. In real energy storage modules several capacitors are typically included and there is a need for high-transconductance switches to balance charging and discharging. An OECT-based differential amplifier was successfully manufactured to balance and limit the voltage across capacitors to 1 V.[140]

Memory and neuromorphic devices

There is a great contemporary interest in networks of devices that exhibit co-location of computation and memory, as these would be extremely efficient at learning specific tasks such as pattern recognition.[144–146] Such systems are termed neuromorphic because they mimic the architecture and/or the function of the nervous system. They rely on devices that exhibit temporary or permanent changes in electrical properties, thereby simulating short-term or long-term memory. The fact that ions in OECTs can change the electrical state of the channel has not gone unnoticed and a variety of memory and neuromorphic devices have been developed (**Fig. 4e**). An adaptive device based on a polyaniline channel and an electrolyte based on LiCl in poly(ethylene oxide) was described in 2005.[147] Hysteresis during a voltage sweep gave rise to bistable operation, which imparted a dependence of the channel resistance on the history of the device. Programmed states were stable for time-scales on the order of ~1,000 s.[148] The device was further optimized and implemented in a neural network architecture[149] and integrated with polysaccharides.[150] An OECT with a similar architecture using a polythiophene semiconductor was reported in 2013,[151] and its functionality as a memory cell was evaluated.[152] One attractive feature of

neuromorphic devices is their potential for low power consumption per switching event. Devices made using polymer nanofibers exhibited energies as low as 1.23 fJ per synaptic spike when fabricated with 300-nm channels.[153] The first neuromorphic transistor based on a PEDOT:PSS OECT was described in 2015.[17] Neuromorphic functions such as paired-pulse depression, adaptation and dynamic filtering were demonstrated in short order. The retention time was increased from a few seconds to several hours by replacing PEDOT:PSS with a modified polymer that undergoes a conformational change upon charging.[154] A system using multiple gates to modulate a single channel was able to discriminate the orientation of voltage patterns imposed on the gate electrode array, emulating the orientation selectivity of the mammalian visual system.[155] Another system using multiple channels and a single gate emulated homeostatic plasticity functions of the brain.[156] A new architecture for organic neuromorphic devices, built as a combination of a battery[157,158] and an OECT, was recently proposed. The different design of this device allows it to attain over 500 distinct synaptic states in a narrow (<1V) voltage range and ensures that the programmed states change by less than 0.1% over tens of hours.[18] Finally, a ferroelectric polymer layer, coated on the gate electrode, was used to control the redox state of a PEDOT:PSS channel.[159] The polarization switching characteristics of the ferroelectric polymer, which take place as the coercive field is overcome, introduced a bistability, used to implement memory functionality.

Outlook

OECTs afford formidable opportunities for research that is at the intersection of materials science, solid state physics, electrochemistry, and electrical engineering. This is exciting, as it sets the stage for collaborative, interdisciplinary work, but can also be somewhat intimidating to newcomers in the field. Although a model that captures the general physics of OECTs exists, a great deal of work is still required

in order to refine it to better understand and optimize device operation. The lateral motion of ions inside the channel, for example, should be considered in future models. This requires the development of 2D drift/diffusion models with the appropriate boundary conditions at the electrolyte/channel interface. On a more fundamental level, while a great deal is known about electronic charge injection at metal/organic interfaces,[160,161] little is known about ion injection from electrolytes into organic semiconductors. This requires the development of tools that are appropriate for studying the processes of ion injection and transport in the simultaneous presence of electronic charge,[39] and at a microscopic level.[162] Moreover, despite the fact that a lot is known about charge transport in organics,[163,164] transport in OECT channels is expected to be particularly challenging to treat due to the presence of water, high charge densities, and dopants. For example, how does the presence of large amount of water affect the density of states of organic semiconductors? Computational tools capable of handling this question are within reach. Finally, on a phenomenological level, the development of SPICE models will greatly boost the design of OECT-based circuits.[165,166]

In terms of materials for OECTs, although some examples of polymers with performance higher than that of PEDOT:PSS have been reported, we are still at the very early stages of development of structure vs. properties relationships that can guide synthesis. As film structure changes upon immersion and operation in an electrolyte environment, the development and utilization of *in situ* and *operando* measurements that combine structural, spectroscopic, and electrical properties are particularly important.[40,41,162] The particular materials properties deemed most important for a given application depend on the purpose, environment, and/or operational lifetime of the device. For example, stability of materials in electrolytes, particularly aqueous ones for biomedical applications, is a primary consideration. The stability requirement might be relaxed, however, for certain applications such as disposable sensors. Biocompatibility is undoubtedly important for implantable devices, but not at all for memory storage devices. Reversibility of the drain current through repeated gate pulses is highly

desirable for most circuit and sensing applications, although hysteretic behavior through trapping/retention of ions and/or morphology changes in the channel paves the way for applications in memory and neuromorphic devices. Tailored selectivity towards different ions can lead to novel biosensors.

Overall, an important materials consideration critical for all applications stems from the fact that the channel of an OECT has to perform two tasks: Transport electronic charge (as measured by μ) and store ionic charge (as measured by C^*). According to Equation (1), the materials figure-of-merit is the product of electronic carrier mobility and volumetric capacitance. This figure-of-merit for example can distinctly show that PEDOT:PSS with ethylene glycol has a μC^* of 75 *F/(cm·V·s)*,[12] new *p*-type materials such as p(g2T-TT) (Fig. 2e) achieved a μC^* of 228 *F/(cm·V·s)*.[30] In contrast, new *n*-type materials such as p(gNDI-g2T) (Fig. 2f) reach a μC^* of ~0.1 *F/(cm·V·s)*.[28] Decoupling these two factors can be very informative as to why one material outperforms another. For example p(g2T-TT) outpaces PEDOT:PSS largely due to its 6-fold increased volumetric capacitance while maintaining a comparable hole mobility. p(gNDI-g2T), has thus far the highest reported C^* (~400 F/cm^3), however its electron mobility is low (~1x10^{-4} $cm^2/V{\cdot}s$). A rule that seems to emerge from available state-of-the-art materials is that hydrophilicity encourages ion transport and hence is a requirement for successful OECT materials candidates,[9,28,30,39,167] although exceptions to this rule have already been identified,[168] and even P3HT has been recently shown to be a channel material in water-based OECTs.[162] Block copolymers and/or blends of semiconducting with ion conducting polymers have been proposed as a possible means to separately control ionic and electronic transport,[169] and these materials are interesting candidates for OECTs. Finally, small molecules have not yet entered the race for high-performance OECT materials. They are expected to do so soon and play a key role for the development of structure vs. properties relationships.

OECTs have been shown to offer versatile fabrication and relaxed design rules, leading to a variety of form factors compatible with low cost, mechanically flexible, large area, and wearable applications. Advantages such as high transconductance, stability in aqueous electrolytes, cytocompatibility and facile biofunctionalization make them particularly suitable for bioelectronics, where some powerful demonstrations have already been achieved. Materials with tailored biological properties, which offer for example direct wiring with enzymes, can play a big role in developing even better OECTs-based sensors. Moving such sensors from in vitro to in vivo implementation represents the next obvious step. The fact that OECTs can be fabricated from materials that are optically transparent, allow electromechanical activation[170] and drug delivery,[171] pave the road for multimodal tools for cell monitoring. 3D cell culture platforms made of conducting polymers[172] create unique opportunities for biomedical tools that control the mechanical, biochemical and electrical microenvironment while monitoring cell behavior at the same time. Significant opportunities exist in combining the high performance of OECT sensors with high-end silicon electronics through heterogeneous integration. One example would be the development of large multiplexed arrays for electrophysiology, as currently practiced using silicon transistors and metal electrodes.[173] At the same time, simple OECT circuits that interface with sensors for multiplexing, processing and amplification will benefit from a reduced set of materials and will find an important niche in applications where signals are not too fast. Ion gels, such as the ones being used in organic FETs,[174] can render OECTs in a format that is suitable for printed electronic applications. The ability of OECTs to handle large currents creates opportunities in power electronics. Finally, the use of OECTs for neuromorphic computing has greatly accelerated in the last few years and small numbers of devices with promising properties have been demonstrated. The next step will entail scaling up the number of devices and scaling down the size of individual devices in order to demonstrate the potential of OECT-based neuromorphic devices to perform increasingly complex tasks.

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Figures

Fig. 1: (a) Typical OECT architecture showing the various components and the biasing scheme. (b) Transfer curve showing depletion-mode operation of an OECT with a conducting polymer channel. At zero gate voltage, holes on the PEDOT contribute to a high drain current and the transistor is ON. When a gate voltage is applied, the holes are replaced by cations and the transistor is OFF. (c) Transfer curve showing accumulation-mode operation of an OECT with a semiconducting polymer channel. At zero gate voltage, the channel has few mobile holes and the transistor is OFF. When a gate voltage is applied, holes accumulate and compensate injected anions and the transistor is OFF. (d) Ionic and electronic circuits used to model OECTs. (e) Voltage distribution in the ionic circuit for two different ratios of gate and channel capacitances.

Fig. 2: Different classes of materials used in OECT channels including conducting polymers (a, b), conjugated polyelectrolytes (c, d) and semiconducting polymers (e, f).

Fig. 3: OECTs are available on a variety of form factors, including planar devices on rigid substrates (a), devices that are flexible and stretchable (b), on fiber (c), integrated with microfluidics (d) and having channels that are in the form of 3D scaffolds (e).

Fig. 4: OECTs are being developed for applications including electrophysiology (a), impedance sensing (b), analyte detection (c), circuits (d), and neuromorphic systems (e).

Box 1: Three different types of transistors. In an FET (a), the gate and channel form the two plates of a capacitor. The charge induced in the channel is given by Q=CV, where C is the capacitance of the oxide. Its capacitance is inversely proportional to its thickness, hence making the oxide thin in order to induce large amounts of charge. In an electrolyte-gated FET (b) a double layer capacitor is formed at the electrolyte/channel interface and electronic charge in the semiconductor is compensated by ionic charge in the electrolyte. In both of these devices, the doping is localized in a think region near the interface of the channel. In an OECT (c), ions penetrate inside the semiconductor, leading to massive changes in the doping state throughout the channel.